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June 20, 2000

Asst. Commissioner of Patents  
Washington, D.C. 20231

**PATENT APPLICATION TRANSMITTAL LETTER**

Inventor(s): Yukikatsu IMANAKA et al.

SINGLE-LAYER TYPE ELECTROPHOTOSENSITIVE MATERIAL AND  
IMAGE FORMING APPARATUS USING THE SAME

Attorney Docket No.: 32739W029

Sir:

Transmitted herewith for filing are the following:

New patent application including 41 pages of text, 2 sheets of formal drawings,  
signed Declaration, Preliminary Amendment, Recordation Cover Sheet and signed  
Assignment, and a check for \$730.00.

Counsel's check for the fee which has been calculated as shown below.

Basic Fee	\$ 690.00
Assignment Fee	\$ <u>40.00</u>
<b>TOTAL:</b>	<b>\$ 730.00</b>

If any additional fees associated with this communication are required, please notify  
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Page 2

Respectfully submitted,

Robert G. Weilacher

Reg. No. 20,531

Atty. Dkt. No.  
32739W029

**PATENT**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant(s):

U.S. Serial No.: To Be Assigned

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Filed: : June 20, 2000 (Herewith)

Examiner: To Be Assigned

For : SINGLE-LAYER TYPE ELECTROPHOTOSENSITIVE MATERIAL AND IMAGE  
FORMING APPARATUS USING THE SAME

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Before calculating the filing fee, please amend the application filed herewith as follows:


**IN THE CLAIMS:**

Please amend claim 10 as follows.

Claim 10, page 40, line 14, delete "of claim 4".

Respectfully submitted,

Beveridge, DeGrandi, Weilacher & Young  
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SINGLE-LAYER TYPE ELECTROPHOTOSENSITIVE MATERIAL  
AND IMAGE FORMING APPARATUS USING THE SAME

BACKGROUND OF THE INVENTION

5           The present invention relates to a single-layer type electrophotosensitive material which is used in digital image forming apparatuses such as electrophotosensitive copying machine, facsimile and laser beam printer, and a digital image forming method using the same.

10           More particularly, the present invention relates to a single-layer type electrophotosensitive material, which does not generate a memory image even when using in a reversal development type digital image forming apparatus including no charge neutralizing step, and a  
15 reversal development type digital image forming method using the same, which does not include a charge neutralizing step.

20           Recently, an organic photosensitive material has widely been used because of its easy production, low cost, wide range of choice of photosensitive materials such as electric charge transferring material, electric charge generating material and binder resin, and high functional design freedom as compared with a conventional inorganic photosensitive material.

25           The organic photosensitive material includes, for

example, single-layer type photosensitive material wherein an electric charge transferring material (hole transferring material, electron transferring material) is dispersed in the same photosensitive layer, together with an electric charge generating material, and multi-layer type photosensitive material comprising an electric charge generating layer containing an electric charge generating material and an electric charge transferring layer containing an electric charge transferring material, which are mutually laminated. The single-layer type photosensitive material has attracted special interest recently because it has such an advantage that optical characteristics can be improved due to simple layer structure, excellent productivity and less interfaces between layers.

On the other hand, an image forming apparatus using an electrophotosensitive system is capable of charging a photosensitive material (principal charge step), exposing an image to form a static latent image (exposure step), developing the static latent image with a toner at a state where a developing bias voltage is applied (development step), transferring the formed toner image to a transfer paper (transfer step), and fixing to form an image. The residual toner on the photosensitive material is cleaned by a urethane blade (cleaning step)

and the residual electric charges on the photosensitive material are neutralized by LED (charge neutralizing step).

To reduce the size of the image forming apparatus and initial cost, various trials of omitting the cleaning step and charge neutralizing step have been made.

The image forming apparatus using an electrophotosensitive system includes, for example, digital and analogue copying machines, facsimile and laser beam printer. In particular, a reversal development system for developing using a toner having the same polarity as that of a charging voltage to be applied to the photosensitive material in the charge step has widely been used in a digital image forming apparatus.

However, the following problems such as generation of a memory image occur when using a conventional electrophotosensitive material in a reversal development type digital image forming apparatus.

<Transfer memory> When using an electrophotosensitive material in a reversal development type digital image forming apparatus, a transfer voltage to be applied to the electrophotosensitive material in a transfer step is usually applied through a transfer medium (paper) without being applied directly to the electrophotosensitive

material, and the transfer voltage is not applied when the transfer medium does not pass through the transfer step.

However, on-off timing of the transfer voltage is very difficult and portion to be applied directly to the photosensitive material is often generated with respect to front/rear portions of the transfer medium. That is, application of the transfer voltage starts before a transferring apparatus is covered with the front portion of the transfer medium. Furthermore, the transfer voltage is continuously applied even if portion of the transferring apparatus is exposed by passage of the rear end of the transfer medium so that the transfer voltage is applied directly to the photosensitive material at said portion.

In case of a positively charging single-layer type photosensitive material, since the polarity of the voltage to be applied in the transferring apparatus is negative, negative space electric charges are remained at the portion of the photosensitive material to which a negative voltage has been applied. Generally, a single-layer type photosensitive material has sensitivity in both polarities so that negative space electric charges are neutralized in the following charge neutralizing step.



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charge step and the memory image is liable to be generated in case where the sensitivity of the positively charging single-layer type photosensitive material to the negative polarity is inferior and the photosensitive layer is used in the image forming apparatus including no charge neutralizing step.

#### SUMMARY OF THE INVENTION

Thus, an object of the present invention is to provide a single-layer type electrophotosensitive material, which hardly generates a exposure memory and a transfer memory and does not generate a memory image even when using in a reversal development type image forming apparatus including no charge neutralizing step.

Another object of the present invention is to provide a reversal development type digital image forming apparatus using the single-layer type electrophotosensitive material, which does not include a charge neutralizing step

The present inventors have intensively studied to attain the above objects and found the fact that a single-layer type electrophotosensitive material comprising a conductive substrate and a photosensitive layer formed on the conductive substrate, characterized in that the photosensitive layer contains a phthalocyanine compound as an electric charge generating

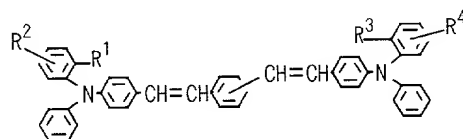
material, a hole transferring material and an electron transferring material in a binder resin, and that a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity measured under the conditions of an exposure wavelength of 780 nm and an exposure energy of  $1.0 \mu\text{J}/\text{cm}^2$  is not more than 500 V hardly generates an exposure memory and a transfer memory and does not generate a memory image even when using in a reversal development type image forming apparatus including no charge neutralizing step. They have further studied based on this finding, thus completing the present invention.

The present invention includes the following inventions:

- (1) a single-layer type electrophotosensitive material comprising a conductive substrate and a photosensitive layer formed on the conductive substrate, wherein the photosensitive layer contains a phthalocyanine compound as an electric charge generating material, a hole transferring material and an electron transferring material in a binder resin, and a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity measured under the conditions of an exposure wavelength of 780 nm and an exposure energy of  $1.0 \mu\text{J}/\text{cm}^2$  is not more than 500 V;

(2) The single-layer type electrophotosensitive material according to the term (1), wherein the absolute value of the plus polarity sensitivity is smaller than that of the minus polarity sensitivity;

5 (3) The single-layer type electrophotosensitive material according to the term (2), which contains, as the hole transferring material, a compound represented by the general formula (1):



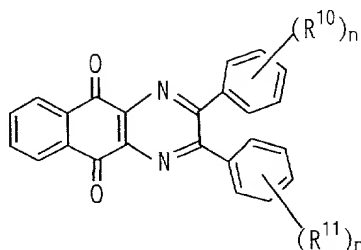
10 wherein  $R^1$  and  $R^3$  are the same or different and each represents an alkyl group which may have a substituent, an aryl group which may have a substituent, or an aralkyl or alkoxy group which may have a substituent; and  $R^2$  and  $R^4$  are the same or different and each represents a hydrogen atom, or an alkyl or alkoxy group which may have a substituent, provided that  $R^2$  and  $R^4$  are hydrogen atoms when  $R^2$  and  $R^4$  are substituted at the para-position;

20 (4) The single-layer type electrophotosensitive material according to the term (2), which contains, as the electron transferring material, at least one selected from the group of compounds represented by the general formula (2):

O=C1C(=O)c2ccccc2C1Oc3cc(R9a)cc(R9b)cc3Oc4cc(R9c)cc(R9d)cc4

wherein  $R^{9a}$ ,  $R^{9b}$ ,  $R^{9c}$  and  $R^{9d}$  are the same or different

and each represents a hydrogen atom, or an alkyl or aryl group which may have a substituent; and the general formula (5):



5 wherein  $R^{10}$  and  $R^{11}$  are the same or different and each represents an alkyl group, a halogenated alkyl group, an aryl group, an aralkyl group, an alkoxy group, an aryloxy group, an aralkyloxy group, an acyl group, an alkoxy carbonyl group, an aryloxy carbonyl group, an aralkyloxy carbonyl group, or a nitro group; and  $n$  represents an integer of 0 to 3;

(5) The single-layer type electrophotosensitive material according to the term (2), which contains, as the hole transferring material, a compound represented by the general formula (1) and, as the electron transferring material, a compound represented by the general formula (2).

(6) The single-layer type electrophotosensitive material according to the term (2), wherein the content of the phthalocyanine compound is from 0.1 to 4.0% by weight based on the weight of the binder resin;

(7) The single-layer type electrophotosensitive

material according to the term (2), which contains, as the binder resin, a bisphenol Z type polycarbonate resin having a weight-average molecular weight of 15,000 to 100,000;

5 (8) The single-layer type electrophotosensitive material according to the term (2), wherein the film thickness of the photosensitive layer is from 10 to 35  $\mu\text{m}$ ;

10 (9) A method of producing a single-layer type electrophotosensitive material comprising a conductive substrate and a photosensitive layer formed on the conductive substrate, the photosensitive layer containing a phthalocyanine compound as an electric charge generating material, a hole transferring material and an electron transferring material in a binder resin, 15 wherein the photosensitive layer is formed by selecting the phthalocyanine compound, hole transferring material, electron transferring material and binder resin so that a difference in absolute value between a plus polarity 20 sensitivity and a minus polarity sensitivity is not more than 500 V under the measuring conditions of an exposure wavelength of 780 nm and an exposure energy of 1.0  $\mu\text{J}/\text{cm}^2$ .

(10) The method of producing a single-layer type 25 electrophotosensitive material according to the term (9),

wherein at least one selected from the group of the compounds represented by the general formulas (2), (3), (4) and (5) of the term (4) is contained as the electron transferring material; and

5 (11) A reversal development type digital image forming apparatus using the single-layer type electrophotosensitive material of the term (1), comprising at least a principal charge step, an exposure step, a development step and a transfer step along the  
10 forward direction of the electrophotosensitive material, wherein a voltage to be applied in the transfer step has a polarity reverse to a voltage to be applied in the charge step.

In the present invention, as described above, when  
15 using an electrophotosensitive material wherein a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity measured under the conditions of an exposure wavelength of 780 nm and an exposure energy of  $1.0 \mu\text{J}/\text{cm}^2$  is not more than  
20 500 V, an exposure memory and a transfer memory are drastically reduced. The reason is considered as follows. That is, the smaller a difference in absolute value of the sensitivity between a plus polarity and a minus polarity, the better the transferring balance  
25 between holes and electrons generated in the



photosensitive layer. Thus, the memory is reduced.

As described above, the absolute value of the plus polarity sensitivity is smaller than that of the minus polarity sensitivity, that is, a positively charging type electrophotosensitive material is the most common because design of an electron transferring material having large mobility is difficult and the mobility of the electron transferring material is smaller than that of the hole transferring material and, furthermore, ozone is hardly generated in the image forming apparatus in the above single-layer type electrophotosensitive material.

The single-layer type electrophotosensitive material according to the present invention preferably includes a positively charged type one as referred to in the above terms (3) to (8).

In the present invention, the positively charging single-layer type electrophotosensitive material preferably contains the compound represented by the general formula (1) as the hole transferring material and at least one of the compounds represented by the general formulas (2), (3), (4) and (5) as the electron transferring material. The reason is as follows. That is, by using the compound having high hole transferability or electron transferability, the

sensitivity to the positive or negative polarity is improved, which is very effective to reduce the memory.

As described in the term (6), even when using in a reversal development type digital image forming apparatus including no charge neutralizing step, the memory image is not generated because the single-layer type electrophotosensitive material of the present invention has a small memory.

As described above, even if the single-layer type electrophotosensitive material of the present invention is used in a reversal development type digital image forming apparatus including no charge neutralizing step, a memory image is not generated because of very small exposure memory and transfer memory.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a graph showing a relationship between a transfer memory potential and an exposure memory potential, and a difference in absolute value of sensitivity between a plus polarity and a minus polarity of single-layer type electrophotosensitive materials of Examples and Comparative Examples.

Fig. 2 is a diagram showing an original for evaluation of a transfer memory image, and a transfer memory image.

Fig. 3 is a diagram showing an original for

evaluation of an exposure memory image, and an exposure memory image.

#### DISCLOSURE OF THE INVENTION

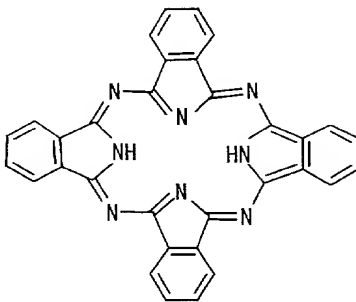
Various materials used in the single-layer type  
5 electrophotosensitive material of the present invention will be described in detail hereinafter.

<Electric charge generating agent> When using laser  
as a light source in a digital image forming apparatus,  
a semiconductor laser and LED are exclusively used in  
10 view of small size, cheap price and simplicity.  
Accordingly, an organic photosensitive material having  
sensitivity in a wavelength range from 700 to 850 nm is  
required. As the electric charge generating material  
which satisfies the above requirement and used in the  
15 organic photosensitive material, for example,  
polycyclic quinone compound, pyrylium compound,  
squalium compound, phthalocyanine compound and azo  
compound have been suggested or put into practice. In  
the single-layer type electrophotosensitive material of  
20 the present invention, various phthalocyanine compounds  
are used.

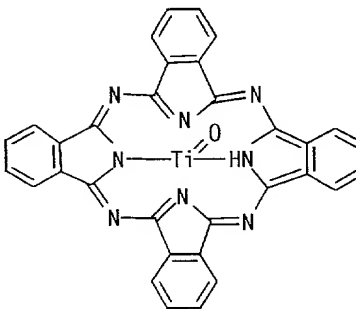
In general, the phthalocyanine compound includes,  
for example, metal-free phthalocyanine (CGM-1)  
containing no center metal; titanyl phthalocyanine  
25 (CGM-2) which has intensively been developed, recently;

and metal phthalocyanine containing a center metal, such as aluminum phthalocyanine, vanadium phthalocyanine, cadmium phthalocyanine, antimony phthalocyanine, chromium phthalocyanine, copper 4-phthalocyanine, germanium phthalocyanine, iron phthalocyanine, chloroaluminum phthalocyanine, chloroindium phthalocyanine, chlorogallium phthalocyanine, magnesium phthalocyanine, dialkyl phthalocyanine, tetramethyl phthalocyanine, and tetraphenyl phthalocyanine. The crystal form that can be used may be any of  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\delta$ ,  $\varepsilon$ ,  $\sigma$ ,  $\chi$  and  $\tau$  forms.

<CGM-1>



<CGM-2>



The phthalocyanine compound is preferably contained in the amount of 0.1 to 4.0% by weight based

on the weight of the binder resin. When the content of the phthalocyanine compound exceeds 4% by weight, a memory becomes larger. The reason is as follows. That is, since carries are incorporated into a trap of the photosensitive layer to generate residual carries as a memory, the number of residual carriers are increased due to excess number of carrier. On the other hand, when the content is less than 0.1% by weight, it becomes difficult to put into practice because of poor photosensitivity.

<Hole transferring material> As the hole transferring material used in the single-layer type electrophotosensitive material of the present invention, a stilbene compound represented by the general formula (1) can be used particularly preferably. When using the stilbene compound as the hole transferring material, the stilbene compound may be contained alone or at least one of them may be contained. That is, various hole transferring materials may be contained, together with the stilbene compound.

Various hole transferring materials include nitrogen-containing cyclic compounds, for example, oxadiazole compound such as 2,5-di(4-methylaminophenyl)-1,3,4-oxadiazole, styryl compound such as 9-4-(diethylaminostyryl)anthracene,

carbazole compound such as polyvinylcarbazole, organic polysilane compound, pyrazoline compound such as 1-phenyl-3(p-dimethylaminophenyl)pyrazoline, hydrazone compound, triphenylamine compound, indole  
5 compound, oxadiazole compound, isoxazole compound, thiazole compound, thiadiazole compound, imidazole compound, pyrazole compound, and triazole compound.

The content of the hole transferring material is preferably from 5 to 500% by weight, and more preferably  
10 from 25 to 200% by weight, based on the weight of the binder resin.

<Electron transferring material> As the electron transferring material used in the single-layer type electrophotosensitive material of the present invention,  
15 a quinone compound represented by the general formula (2), (3), (4) or (5) can be used particularly preferably. When using the quinone compound as the electron transferring material, the quinone compound may be contained alone or at least one of them may be contained.  
20 That is, other electron transferring materials may be contained, together with the quinone compound.

Other electron transferring materials include electron attractive substances, for example, pyrene compound, carbazole compound, hydrazone compound,  
25 N,N-dialkylaniline compound, diphenylamine compound,

triphenylamine compound, triphenylmethane compound,  
tetracyanoethyl, tetracyanoquinoedimethane,  
chloroanil, bromoanil, 2,4,7-trinitro-9-fluorenone,  
2,4,5,7-tetranitro-9-fluorenone,  
5 2,4,7-trinitro-9-dicyanomethylenefluorenone,  
2,4,5,7-tetranitroxanthone, and  
2,4,8-trinitrothioxanthone, or those prepared by  
polymerizing these electron attractive substances.

The content of the electron transferring material  
10 is preferably from 5 to 100% by weight, and more preferably  
from 10 to 80% by weight, based on the weight of the binder  
resin.

<Binder resin> As the binder resin in which the above  
respective components are dispersed, for example, there  
15 can be used various resins used conventionally in the  
photosensitive layer.

There can be used, for example, thermoplastic resins  
such as styrene-butadiene copolymer,  
styrene-acrylonitrile copolymer, styrene-maleic acid  
20 copolymer, acrylic copolymer, styrene-acrylic acid  
copolymer, polyethylene, ethylene-vinyl acetate  
copolymer, chlorinated polyethylene, polyvinyl chloride,  
polypropylene, ionomer, vinyl chloride-vinyl acetate  
copolymer, alkyd resin, polyamide, polyurethane,  
25 polycarbonate, polyacrylate, polysulfone, diallyl

phthalate resin, ketone resin, polyvinyl butyral resin, and polyether resin; crosslinkable thermosetting resins such as silicone resin, epoxy resin, phenol resin, urea resin, and melamine resin; and photocurable resins such as epoxy acrylate and urethane acrylate. These binder resins can be used alone, or two or more kinds of them can be used in combination.

Particularly preferred resin includes, for example, bisphenol Z type monomer and bisphenol Z type polycarbonate derived from phosgene, such as Panlight manufactured by Teijin Chemicals Co., Ltd. and PCZ manufactured by Mitsubishi Gas Chemicals Co., Ltd.

The weight-average molecular weight of the binder resin is preferably within a range from 15,000 to 100,000.

In addition to the above respective components, various conventionally known additives such as antioxidants, radical scavengers, singlet quenchers, deterioration inhibitors (e.g. ultraviolet absorbers), softeners, plasticizers, surface modifiers, extenders, thickeners, dispersion stabilizers, waxes, acceptors, and donors can be incorporated into the single-layer type electrophotosensitive material of the present invention as far as these additives do not exert a deleterious influence on electrophotosensitive characteristics. To improve the sensitivity of the photosensitive layer,



for example, known sensitizers such as terphenyl, halonaphthoquinones, and acenaphthylene may be used in combination with the electric charge generating material.

5        In the single-layer type electrophotosensitive material, a barrier layer may be formed between the conductive substrate and photosensitive layer as far as it does not inhibits the characteristics of the photosensitive material.

10        In the single-layer type electrophotosensitive material of the present invention, the film thickness of the photosensitive layer is preferably within a range from about 10 to 35  $\mu$ m. When the film thickness exceeds 35  $\mu$ m, the memory becomes large. The reason is  
15        considered as follows. That is, as the film thickness of the photosensitive layer increases, dark decay increases to reduce a charging capability, whereby an influence of the memory is liable to be exerted. Alternatively, a trap increase by an increase in absolute  
20        quantity of the constituting materials of the photosensitive layer. On the other hand, when the film thickness is less than 10  $\mu$ m, the sensitivity is drastically lowered by removal of the film, thereby making it difficult to put into practice.

25        As described hereinbefore, the single-layer type

electrophotosensitive material according to the present invention includes a preferable embodiment which contains a compound represented by the general formula (1) as the hole transferring material and a compound represented by the general formula (2) as the electron transferring material. Especially, it is more preferable to select HTM-1 as a compound represented by the general formula (1) and ETM-1 as a compound represented by the general formula (2).

The single-layer type electrophotosensitive material comprises a conductive substrate and a single photosensitive layer formed on the conductive substrate. This photosensitive layer is formed by dissolving or dispersing the electric charge generating material, hole transferring material, electron transferring material and binder resin in a proper solvent, coating the conductive substrate with the resulting coating solution and drying the coating solution.

As the conductive substrate on which the photosensitive layer is formed, for example, various materials having the conductivity can be used. Examples thereof include metallic simple substances such as iron, aluminum, copper, tin, platinum, silver, vanadium, molybdenum, chromium, cadmium, titanium, nickel, palladium, indium, stainless steel, and brass; plastic

materials prepared by depositing or laminating the above metal; and glasses coated with aluminum iodide, tin oxide, and indium oxide.

The conductive substrate may be in the form of a sheet or drum according to the structure of the image forming apparatus to be used. The substrate itself may have the conductivity, or the surface of the substrate may have the conductivity. The conductive substrate may be preferably those having a sufficient mechanical strength on use.

When the photosensitive layer is formed by the coating method, a dispersion is prepared by dispersing and mixing the above hole transferring material, electric charge generating material, electron acceptor and binder resin, together with a proper solvent, using a known method such as roll mill, ball mill, attritor, paint shaker, and ultrasonic dispersing equipment, and then the resulting dispersion is coated by using a known means and dried.

As the solvent for preparing the dispersion, various organic solvents can be used. The organic solvent includes, for example, alcohols such as methanol, ethanol, isopropanol, and butanol; aliphatic hydrocarbons such as n-hexane, octane, and cyclohexane; aromatic hydrocarbons such as benzene, toluene, and xylene;

halogenated hydrocarbons such as dichloromethane, dichloroethane, chloroform, carbon tetrachloride, and chlorobenzene; ethers such as dimethyl ether, diethyl ether, tetrahydrofuran, ethylene glycol dimethyl ether, and diethylene glycol dimethyl ether; ketones such as acetone, methyl ethyl ketone, and cyclohexanone; esters such as ethyl acetate and methyl acetate; and dimethylformaldehyde, dimethylformamide, and dimethyl sulfoxide. These solvents can be used alone, or two or more kinds of them can be used in combination.

To improve the dispersion properties of the hole transferring material, electric charge generating material and electron acceptor, and the smoothness of the surface of the photosensitive layer, for example, surfactants and leveling agents may be used.

On the other hand, the image forming apparatus of the present invention is a reversal development type digital image forming apparatus using the single-layer type electrophotosensitive material of the term (1), comprising at least a principal charge step, an exposure step, a development step and a transfer step along the forward direction of the electrophotosensitive material, characterized in that a voltage to be applied in the transfer step has a polarity reverse to a voltage to be applied in the charge step. Examples of the image forming

apparatus include digital copying machine, facsimile and laser beam printer.

Even if the single-layer type electrophotosensitive material of the present invention is used in the above image forming apparatus including no charge neutralizing step, no memory image is generated because of very small transfer and exposure memories.

As described above, the cleaning step may be omitted sometimes, similar to the charge neutralizing step, in order to reduce the size of the image forming apparatus and initial cost.

#### EXAMPLES

The following Examples and Comparative Examples further illustrate the present invention in detail. The following embodiments are illustrative, and they should not be construed to limit the technical scope of the present invention.

#### EXAMPLES 1 to 8

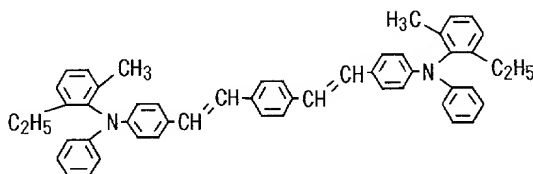
2.0 Parts by weight of a X type metal-free phthalocyanine (CGM-1) as the electric charge generating material, 70 parts by weight of a hole transferring material (HTM-1) represented by the general formula (1), 40 parts by weight of electron transferring materials (ETM-1 to ETM-8) represented by the general formulas (2), (3), (4) and (5), 100 parts by weight a bis-Z type

polycarbonate resin having a weight-average molecular weight of 30,000 as the binder resin and 800 parts by weight of tetrahydrofuran were dispersed or dissolved in a ball mill for 24 hours to prepare a coating solution for single-layer type photosensitive layer. Then, an alumina tube as the substrate was coated with the coating solution according to a dip coating method, followed by hot-air drying at 125 °C for 30 minutes to form a single-layer type photosensitive material having a photosensitive layer of 20  $\mu\text{m}$  in a film thickness.

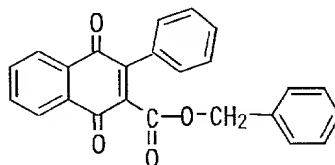
#### COMPARATIVE EXAMPLES 1 to 3

In the same manner as in Examples 1 to 7, except that ETM-9 to ETM-11 were used as the electron transferring material, single-layer type photosensitive materials were produced.

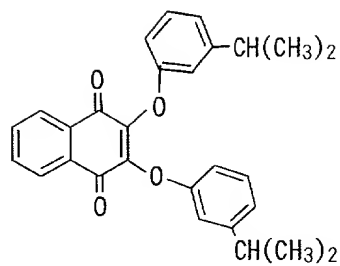
#### <HTM-1>



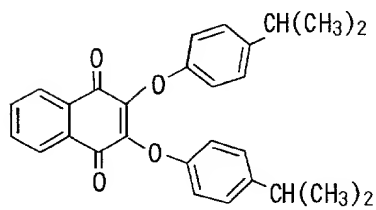
#### <ETM-1>



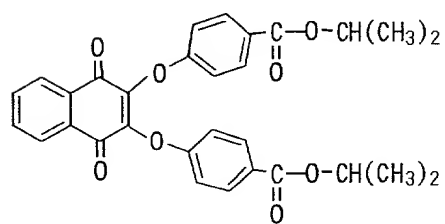
#### 20 <ETM-2>



&lt;ETM-3&gt;

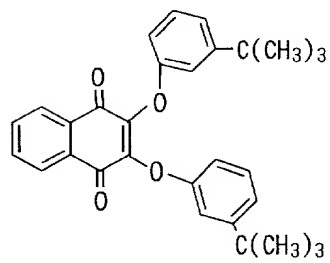


&lt;ETM-4&gt;

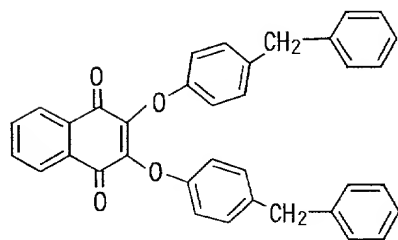


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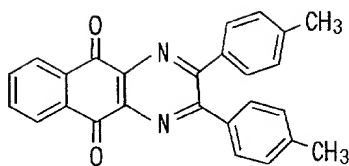
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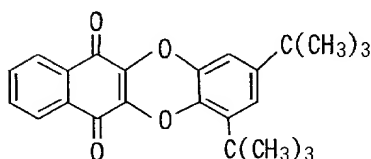
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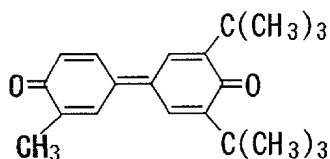
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&lt;ETM-8&gt;

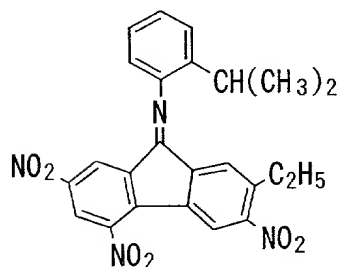


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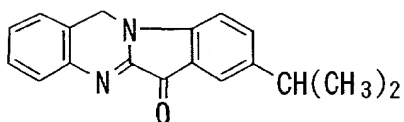


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&lt;ETM-10&gt;



&lt;ETM-11&gt;



10 EXAMPLES 9 to 16

In the same manner as in Examples 1 to 8, except that titanyl phthalocyanine (CGM-2) was used as the electric charge generating material, single-layer type photosensitive materials were produced.

15 COMPARATIVE EXAMPLES 4 to 6



In the same manner as in Comparative Examples 1 to 3, except that titanyl phthalocyanine (CGM-2) was used as the electric charge generating material, single-layer type photosensitive materials were produced.

5 With respect to the single-layer type photosensitive materials of the respective Examples and Comparative Examples, the following respective characteristics were evaluated. The evaluation results are shown in Tables 1 and 2. Among these data, a  
10 relationship between a transfer memory potential, an exposure memory potential, and a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity is shown in Fig. 1.

<Evaluation of plus polarity sensitivity> Using a drum  
15 sensitivity tester (manufactured by GENTEC Co. under the trade name of GENTEC SINCIA 30 M)), a voltage was applied to the electrophotosensitive materials of the respective Examples and Comparative Examples to charge the surface at +800 V. Then, the surface of each photosensitive  
20 material (exposure time: 100 msec.) was irradiated (exposure energy:  $1.0 \mu\text{J}/\text{cm}^2$ ) with monochromatic light having a wavelength of 780 nm (half-width: 20 nm, light intensity:  $20 \mu\text{W}/\text{cm}^2$ ) from white light of a halogen lamp as an exposure light source through a band-pass filter,  
25 and then a surface potential at the time at which 500

msec. have passed since the beginning of exposure was measured as a potential after exposure  $V_{LP}$  (V). The smaller the potential after exposure, the higher the sensitivity of the photosensitive material.

5 [Evaluation of minus polarity sensitivity]

In the same manner as in case of the <evaluation of plus polarity sensitivity>, except that a voltage was applied to the electrophotosensitive materials of the respective Examples and Comparative Examples to charge  
10 the surface at -800 V using a drum sensitivity tester (manufactured by GENTEC Co. under the trade name of GENTEC SINCIA 30 M)), a surface potential at the time at which 500 msec. have passed since the beginning of exposure was measured as a potential after exposure  $V_{LN}$  (V).

15 <Evaluation of transfer memory potential> After the electrophotosensitive materials of the respective Examples and Comparative Examples were installed in a multifunction printer Antico 40 excluding a charge neutralizing lamp, manufactured by KYOCERA-MITA Co.,  
20 Ltd., a surface potential on application of no transfer bias and a surface potential on application of a transfer bias after the following charge step were measured and a difference between them was taken as a transfer memory potential. The case where the transfer memory potential  
25 is 45 V or less at which no transfer memory image is

generated was rated "Pass", whereas, the case where the transfer memory potential is 45 V or more was rated "Fail".

<Evaluation of exposure memory potential> After the electrophotosensitive materials of the respective

5 Examples and Comparative Examples were installed in a multifunction printer Antico 40 excluding a charge neutralizing lamp, manufactured by KYOCERA-MITA Co., Ltd., a surface potential on no exposure and a surface potential on exposure after the following charge step  
10 were measured and a difference between them was taken as an exposure memory potential. Similar to the case of the transfer memory potential, the case where the exposure memory potential is 45 V or less at which no transfer memory image is generated was rated "Pass",  
15 whereas, the case where the exposure memory potential is 45 V or more was rated "Fail".

<Evaluation of transfer memory image> After the electrophotosensitive materials of the respective Examples and Comparative Examples were installed in a  
20 multifunction printer Antico 40 excluding a charge neutralizing lamp, manufactured by KYOCERA-MITA Co., Ltd., a printing test was carried out and it was visually judged whether a transfer memory image is generated or not. As shown in Fig. 2, the transfer memory image refers  
25 to an image wherein a black lateral band was generated

in a drum longitudinal direction by a reduction in surface potential of the photosensitive material at the portion to which the transfer bias was applied in case where the printing test was carried out using an original having a gray front surface (Munsell value:  $N = 6.5$ ).

<Evaluation of exposure memory image> After the electrophotosensitive materials of the respective Examples and Comparative Examples were installed in a multifunction printer Antico 40 excluding a charge neutralizing lamp, manufactured by KYOCERA-MITA Co., Ltd., a printing test was carried out and it was visually judged whether an exposure memory image is generated or not. The exposure memory image refers to an image wherein a ghost image of the exposed portion was generated at the gray portion by a reduction in surface potential of the photosensitive material at the strongly exposed portion (black solid portion) in case where the printing test was carried out using an original as shown in Fig. 3.

[Table 1]

	Kind of ETM	Plus polarity sensitivity (V)	Minus polarity sensitivity (V)	Difference in absolute value of sensitivity (V)	Transfer memory potential (V)	Exposure memory potential (V)	Transfer memory image	Exposure memory potential (V)
Example 1	ET-1	132	150	18	15	10	No memory image was generated	No memory image was generated
Example 2	ET-2	141	290	149	20	26	No memory image was generated	No memory image was generated
Example 3	ET-3	144	341	197	30	32	No memory image was generated	No memory image was generated
Example 4	ET-4	150	352	202	26	10	No memory image was generated	No memory image was generated
Example 5	ET-5	131	153	22	15	25	No memory image was generated	No memory image was generated
Example 6	ET-6	205	322	117	35	32	No memory image was generated	No memory image was generated
Example 7	ET-7	252	550	298	35	36	No memory image was generated	No memory image was generated
Example 8	ET-8	223	704	481	25	44	No memory image was generated	No memory image was generated
Comp. Example 1	ET-9	131	652	521	55	66	Lateral black band was generated	Ghost image was generated
Comp. Example 2	ET-10	123	663	540	70	76	Lateral black band was generated	Ghost image was generated
Comp. Example 3	ET-11	130	661	531	85	90	Lateral black band was generated	Ghost image was generated



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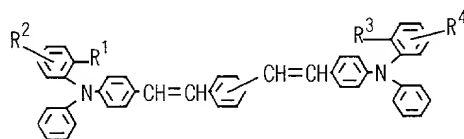
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What is claimed is:

1. A single-layer type electrophotosensitive material comprising a conductive substrate and a photosensitive layer formed on the conductive substrate, characterized in that the photosensitive layer contains a phthalocyanine compound as an electric charge generating material, a hole transferring material and an electron transferring material in a binder resin, and that a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity measured under the conditions of an exposure wavelength of 780 nm and an exposure energy of  $1.0 \mu\text{J}/\text{cm}^2$  is not more than 500 V.

2. The single-layer type electrophotosensitive material according to claim 1, wherein the absolute value of the plus polarity sensitivity is smaller than that of the minus polarity sensitivity.

3. The single-layer type electrophotosensitive material according to claim 2, which contains, as the hole transferring material, a compound represented by the general formula (1):

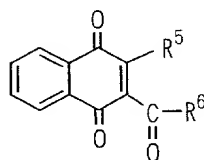


wherein  $R^1$  and  $R^3$  are the same or different and each

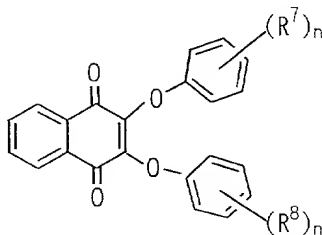


represents an alkyl group which may have a substituent,  
 an aryl group which may have a substituent, or an aralkyl  
 or alkoxy group which may have a substituent; and  $R^2$  and  
 $R^4$  are the same or different and each represents a hydrogen  
 atom, or an alkyl or alkoxy group which may have a  
 substituent, provided that  $R^2$  and  $R^4$  are hydrogen atoms  
 when  $R^2$  and  $R^4$  are substituted at the para-position.

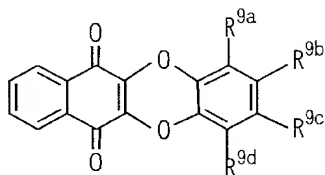
4. The single-layer type electrophotosensitive  
 material according to claim 2, which contains, as the  
 electron transferring material, at least one selected  
 from the group of compounds represented by the general  
 formula (2):



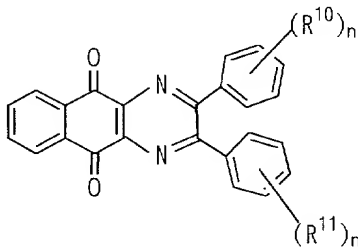
wherein  $R^5$  represents a halogen atom, or an alkyl or aryl  
 group which may have a substituent; and  $R^6$  represents  
 an alkyl or alkoxy group which may have a substituent,  
 or a group:  $-O-R^{6a}$ , which represents an alkyl or aryl  
 group which may have a substituent;  
 the general formula (3):



wherein  $R^7$  and  $R^8$  are the same or different and each represents an alkyl group, a halogenated alkyl group, an aryl group, an aralkyl group, an alkoxy group, an aryloxy group, an aralkyloxy group, an acyl group, an alkoxy carbonyl group, an aryloxy carbonyl group, an aralkyloxy carbonyl group, or a nitro group; and  $n$  represents an integer of 0 to 3; the general formula (4):



wherein  $R^{9a}$ ,  $R^{9b}$ ,  $R^{9c}$  and  $R^{9d}$  are the same or different and each represents a hydrogen atom, or an alkyl or aryl group which may have a substituent; and the general formula (5):



wherein  $R^{10}$  and  $R^{11}$  are the same or different and each represents an alkyl group, a halogenated alkyl group, an aryl group, an aralkyl group, an alkoxy group, an aryloxy group, an aralkyloxy group, an acyl group, an alkoxy carbonyl group, an aryloxy carbonyl group, an aralkyloxy carbonyl group, or a nitro group; and  $n$

represents an integer of 0 to 3.

5        5. The single-layer type electrophotosensitive material according to claim 2, which contains, as the hole transferring material, a compound represented by the general formula (1) and, as the electron transferring material, a compound represented by the general formula (2).

10       6. The single-layer type electrophotosensitive material according to claim 2, wherein the content of the phthalocyanine compound is from 0.1 to 4.0% by weight based on the weight of the binder resin.

15       7. The single-layer type electrophotosensitive material according to claim 2, which contains, as the binder resin, a bisphenol Z type polycarbonate resin having a weight-average molecular weight of 15,000 to 100,000.

      8. The single-layer type electrophotosensitive material according to claim 2, wherein the film thickness of the photosensitive layer is from 10 to 35  $\mu$ m.

20       9. A method of producing a single-layer type electrophotosensitive material comprising a conductive substrate and a photosensitive layer formed on the conductive substrate, the photosensitive layer containing a phthalocyanine compound as an electric  
25       charge generating material, a hole transferring material

and an electron transferring material in a binder resin, characterized in that the photosensitive layer is formed by selecting the phthalocyanine compound, hole transferring material, electron transferring material and binder resin so that a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity is not more than 500 V under the measuring conditions of an exposure wavelength of 780 nm and an exposure energy of  $1.0 \mu\text{J}/\text{cm}^2$ .

10 10. The method of producing a single-layer type electrophotosensitive material according to claim 9, wherein at least one selected from the group of the compounds represented by the general formulas (2), (3), (4) and (5) of claim 4 is contained as the electron  
15 transferring material.

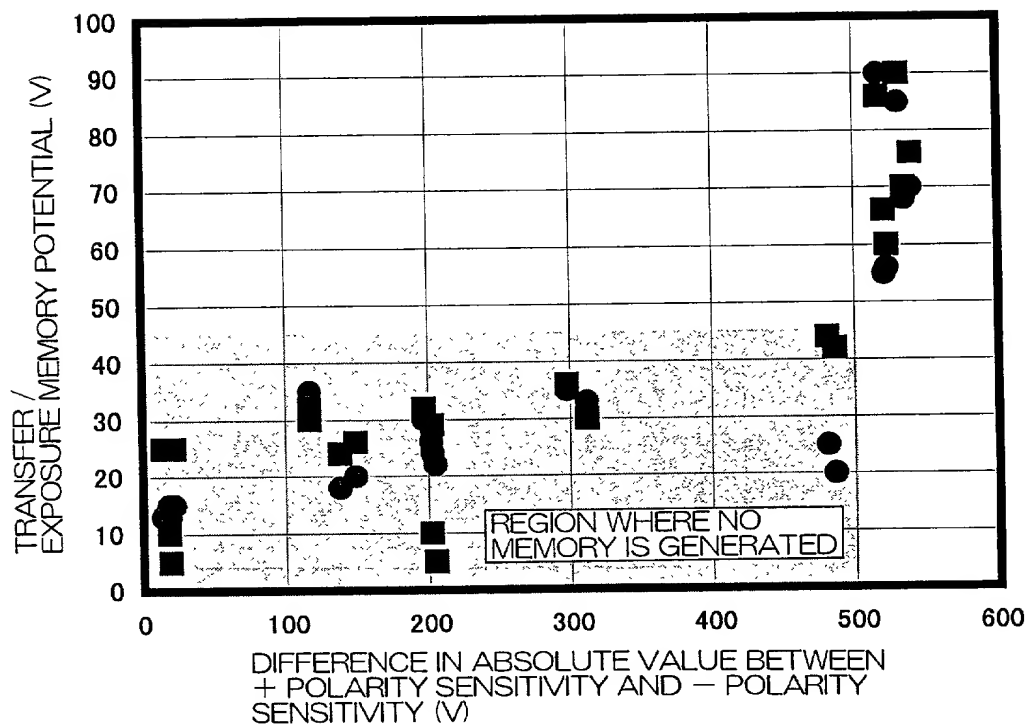
11. A reversal development type digital image forming apparatus using the single-layer type electrophotosensitive material of claim 1, comprising at least a principal charge step, an exposure step, a  
20 development step and a transfer step along the forward direction of the electrophotosensitive material, characterized in that a voltage to be applied in the transfer step has a polarity reverse to a voltage to be applied in the charge step.

## ABSTRACT

The present invention provides a single-layer type electrophotosensitive material comprising a conductive substrate and a photosensitive layer formed on the conductive substrate, wherein the photosensitive layer contains a phthalocyanine compound as an electric charge generating material, a hole transferring material and an electron transferring material in a binder resin, and that a difference in absolute value between a plus polarity sensitivity and a minus polarity sensitivity measured under the conditions of an exposure wavelength of 780 nm and an exposure energy of  $1.0 \mu\text{J}/\text{cm}^2$  is not more than 500 V, and a reversal development type digital image forming apparatus using the electrophotosensitive material, which does not include a charge neutralizing step.

FIG. 1

RELATIONSHIP BETWEEN DIFFERENCE IN  
ABSOLUTION VALUE OF SENSITIVITY AND  
EXPOSURE MEMORY POTENTIAL



● : TRANSFER MEMORY POTENTIAL

■ : EXPOSURE MEMORY POTENTIAL

FIG. 2

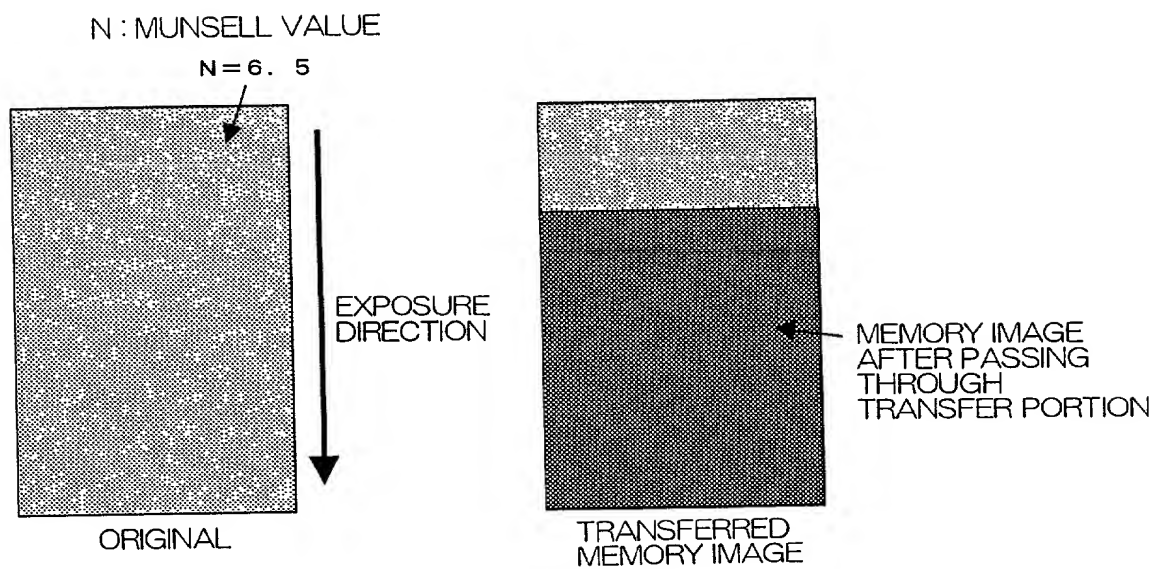
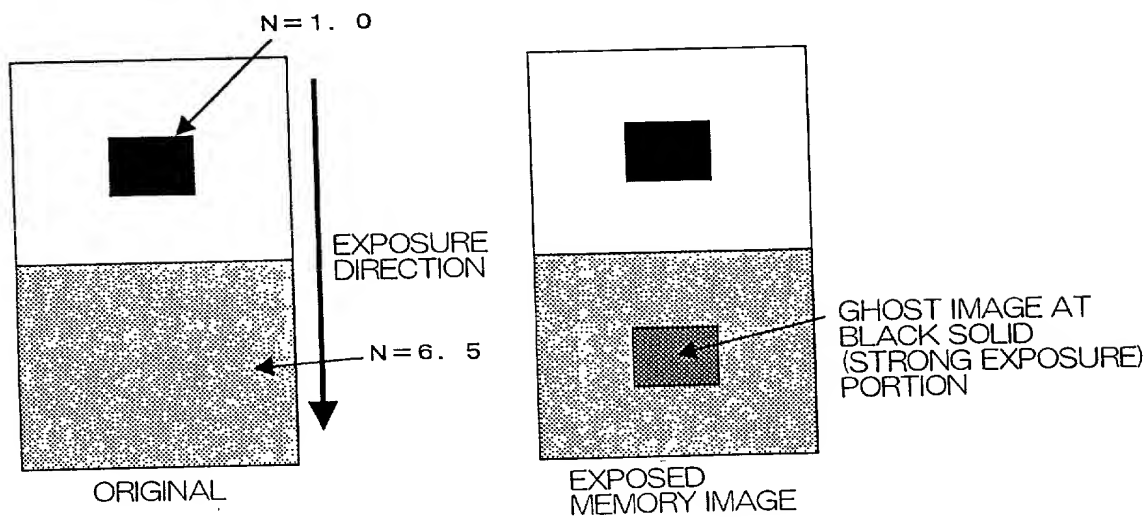


FIG. 3



07/7-1

UNITED STATES (Form BDWY-1)  
 Patents and Design Patents  
 Sole & Joint Inventors  
 Convention & Non-convention  
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 This form cannot be amended, altered  
 or changed after it is signed.  
 (For use only for inventors who  
 understand the English language.)

## Declaration and Power of Attorney United States Patent Application

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

SINGLE-LAYER TYPE ELECTROPHOTOSENSITIVE MATERIAL AND IMAGE FORMING APPARATUS

USING THE SAME, the specification of which  
 (check one) ☒ is attached hereto.

- ☐ was filed as U.S. Application No. \_\_\_\_\_ on \_\_\_\_\_ and (if applicable) was amended on \_\_\_\_\_.
- ☐ was filed as PCT International Application No. \_\_\_\_\_ on \_\_\_\_\_ and (if applicable) was amended under PCT Article 19 on \_\_\_\_\_.

I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) or §365(b) of any foreign and PCT application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States of America listed in this Declaration. I have also identified below any foreign application for patent or inventor's certificate or PCT international application having a filing date before that of the application(s) on which priority is claimed:

Foreign/PCT Application No.	Country	Filing Date	Priority Claimed? (yes/no)
Patent Application No. 11-302914	Japan	October 25, 1999	No

☐ I hereby claim the benefit under Title 35, United States Code, §120 or §365(c) of any United States application and PCT international application designating the United States of America listed in this Declaration and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application or PCT international application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

U.S. Application No.	Filing Date	Status (patented/pending/abandoned?)

I hereby claim priority benefits under Title 35 United States Code §119(e) of any U.S. provisional application(s) listed below:

U.S. Provisional Application No.	Filing Date

I hereby appoint the following attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: Joseph A. DeGrandi (17446), Robert G. Weilacher (20531), Richard G. Young (20628), Michael A. Makuch (32263), Dennis C. Rodgers (32936), Thomas L. Evans (35805), Maurice U. Cahn (30454), Frank C. Cimino, Jr. (39945), and Carolyn A. Favorito (39183).

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Signature: Yukikatsu Imanaka Date: 6 / 7 / 2000  
 (Month/Day/Year)

☒ Additional inventors and/or prior applications are listed in attached Supplemental Sheet(s).

BDWY 1196



# Supplemental Sheet to U.S. Declaration and Power of Attorney

(BDWY-1 supp.)

## Additional Inventor(s):

I hereby declare that all statements made in the attached Declaration and Power of Attorney of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Signature: \_\_\_\_\_ Date: \_\_\_\_\_  
 (Month/Day/Year)

## Additional Foreign/PCT Applications:

Foreign/PCT Application No.	Country	Filing Date	Priority Claimed? (yes/no)

## Additional U.S. Applications:

U.S. Application No.	Filing Date	Status (patented/pending/abandoned?)

## Additional Provisional U.S. Applications:

U.S. Provisional Application No.	Filing Date